PB84-153691

# Aircraft Observations of Regional Transport of Ozone in the Northeastern United States

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14 Feb 83

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| 1. REPORT NO. EPA-600/J-83-149 | 2. PB84-153691 |
| 4. TITLE AND SUBTITLE: AIRCRAFT OBSERVATIONS OF REGIONAL TRANSPORT OF OZONE IN THE NORTHEASTERN UNITED STATES (Journal Version) | 5. REPORT DATE 1983<br>6. PERFORMING ORGANIZATION CODE |
| 7. AUTHOR(S): J. F. Clarke and J. K. S. Ching | 8. PERFORMING ORGANIZATION REPORT NO. |
| 9. PERFORMING ORGANIZATION NAME AND ADDRESS: (Same as Block 12) | 10. PROGRAM ELEMENT NO. CDWA1A/02-1336 (FY-82)<br>11. CONTRACT/GRANT NO. |
| 12. SPONSORING AGENCY NAME AND ADDRESS: Environmental Sciences Research Laboratory--RTP,NC<br>Office of Research and Development<br>U.S. Environmental Protection Agency<br>Research Triangle Park, North Carolina 27711 | 13. TYPE OF REPORT AND PERIOD COVERED<br>14. SPONSORING AGENCY CODE EPA/600/09 |

15. SUPPLEMENTARY NOTES

REF: Atmospheric Environment, 17(9):1703-1712, 1983

16. ABSTRACT

A regional scale aircraft sampling program was conducted during August 1979 to obtain data for validation of a regional scale photochemical air quality simulation model and for studying the physical and chemical processes important in long-range transport of ozone and ozone precursors throughout the diurnal cycle. Three aircraft were used to obtain continuous measurements of $O_3$, NO, $NO_x$, B-scat, and grab samples for later analysis for hydrocarbon species. The field program is described and Lagrangian sampling data for a 24-hour period presented and discussed with the conclusion that relatively high ozone concentrations, generated within urban plumes, were transported from Ohio to the Northeast Corridor.

17. KEY WORDS AND DOCUMENT ANALYSIS

| a. DESCRIPTORS | b. IDENTIFIERS/OPEN ENDED TERMS | c. COSATI Field/Group |
|---|---|---|
| | | |

| | | |
|---|---|---|
| 18. DISTRIBUTION STATEMENT: RELEASE TO PUBLIC | 19. SECURITY CLASS *(This Report)* UNCLASSIFIED | 21. NO. OF PAGES 11 |
| | 20. SECURITY CLASS *(This page)* UNCLASSIFIED | 22. PRICE |

EPA Form 2220-1 (9-73)

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SPRINGFIELD, VA. 22161

EPA-600/J-83-149
JOURNAL ARTICLE

*Atmospheric Environment* Vol. 17, No. 9, pp. 1703–1712, 1983
Printed in Great Britain.

0004-6981/83 $3.00 + 0.00
Pergamon Press Ltd.

# AIRCRAFT OBSERVATIONS OF REGIONAL TRANSPORT OF OZONE IN THE NORTHEASTERN UNITED STATES

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(*First received* 23 *August* 1982 *and received in final form* 14 *February* 1983)

**Abstract**—A regional-scale aircraft sampling program was conducted during August 1979 to obtain data for validation of a regional-scale photochemical air quality simulation model and for studying the physical and chemical processes important in long-range transport of $O_3$ and $O_3$ precursors throughout the diurnal cycle. Three aircraft were deployed in a Lagrangian sampling mode to obtain continuous measurements of $O_3$, oxides of nitrogen, nephelometer scattering coefficient, and grab samples to be analyzed later for hydrocarbon species. The analysis reported herein is for a 24 h period during which relatively high $O_3$ concentrations, generated within urban plumes, were transported from Ohio to the NE Corridor.

## INTRODUCTION

There has been substantial documentation in recent years that tropospheric air containing $O_3$ at concentrations near or above the National Ambient Air Quality Standard (0.12 ppm) may be transported over long distances. Spicer *et al.* (1979) and White *et al.* (1976) showed significant $O_3$ transport in urban plumes extending beyond 250 km downwind from source areas. An analysis of Wolff *et al.* (1977) indicated enhancement of $O_3$ concentrations in the NE Corridor due to long-distance transport. Vukovich *et al.* (1977) presented data which indicate episodes of high $O_3$ concentrations are associated with quasi-stationary high pressure systems, and Samson and Ragland (1977) presented evidence of large-scale transport of $O_3$ within high pressure systems.

Recognizing that regional transport of $O_3$ and $O_3$ precursors must be taken into account in developing control strategies for oxidants in the Northeast, the U.S. Environmental Protection Agency (EPA) initiated a program in 1976 to develop a regional-scale photochemical air quality simulation model. The regional model will provide inflow boundary conditions for urban-scale photochemical models. However, the model is basically applicable to long-distance transport of any pollutant, because it contains, in addition to chemical kinetics, a comprehensive mathematical description of boundary layer and regional-transport processes (see Lamb, 1983). The modeling area, shown in Fig. 1, extends from eastern Ohio to the East Coast and from northern Virginia to southern Maine.

Because of the complexities of the simulations of meteorological dynamics and chemical kinetics, a detailed 3-dimensional network of pollution and meterological data was needed to evaluate the model. To collect these data, EPA conducted the Northeast Regional Oxidant Studies (NEROS) during the summers of 1979 (NEROS I) and 1980 (NEROS II).

NEROS I was conducted during August 1979 to collect chemical species data on a regional scale (by aircraft) for the purpose of development and validation of the regional model. Upper-air soundings were obtained four times daily at most National Weather Service upper-air stations in the modeling area and also at Pennsylvania State University, University Park, PA. $O_3$ surface deposition rates and cloud venting of boundary-layer $O_3$ were studied near Lancaster, PA.

NEROS II was designed primarily to study the fate of $O_3$ produced within urban plumes from their sources to the point at which they blend with the regional background burden. For this purpose, the Columbus, OH and Baltimore, MD, urban plumes were aerially sampled between 14 July and 15 August, 1980. The Columbus NEROS program is described by Vaughan *et al.* (1982).

This paper describes the NEROS I field program. Specifically, a case study, 3–4 August 1979, is presented which demonstrates regional-scale transport of $O_3$. The analysis, based primarily on aircraft measurements, follows an air mass in a Lagrangian mode through a diurnal cycle as it was transported from Ohio to the East Coast.

## EXPERIMENTAL DESIGN

Three aircraft were used in NEROS I to obtain data over the 3-dimensional domain of the NEROS grid for 24- to 48-h periods: a Piper Navajo operated by Research Triangle Institute, an Aero Commander 680 operated by Washington State University, and a Britten Norman Islander operated by the Brookhaven

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National Laboratory. Each aircraft was instrumented to obtain continuous measurement of nitric oxide (NO), total oxides of nitrogens ($NO_x$), ozone ($O_3$), nephelometer scattering coefficient ($\sigma_s$), temperature, relative humidity or dew point, and for taking grab samples to be analyzed later for hydrocarbon species.

Aerial sampling was conducted in a forecast Lagrangian mode, i.e. an initial specified volume of air was sampled at several forecast positions as it was being transported over the NEROS grid. An experimental period consisted of a sampling flight starting at about 1200 EST, to establish upwind boundary conditions (e.g. see track A–B on Fig. 1), and four additional flights at approximately 6-h intervals at downwind traverses specified to approximate Lagrangian sampling of the previous traverse. In the course of a traverse, the aircraft flew 75- to 100-km legs alternately within the afternoon mixed layer (or its residual for nocturnal flights) and above the mixed layer. On some traverses a spiral was made to about 3000 m. This sampling scenario, in addition to providing 3-dimensional validation data for the regional model, afforded the opportunity to study physical and chemical processes associated with Lagrangian $O_3$ transport, both within and above the mixed layer.

The aircraft were directed from a control center operated by Brookhaven National Laboratory, Upton, NY. Meteorological data and predictions were used to establish the orientation and spacing of the flight tracks. Meteorological information was upgraded for each flight, to increase the likelihood that each subsequent flight track represented Lagrangian sampling of the previous track. Six experimental periods were conducted during August 1979.

## CASE STUDY ANALYSIS: 3–4 AUGUST

On 2 August 1979, a weak cold front passed through the Ohio Valley and moved to the East Coast where it became stationary for about 48 h. Surface fronts, isobars, and pressure centers for 3 August are shown in Fig. 1. Boundary layer transport winds behind the cold front were generally westerly and initial conditions for Lagrangian air mass sampling were established by aerial sampling along track A–B on Fig. 1 from 1151 to 1359 EST on 3 August. Based on meteorological data and forecasts obtained over the next 24 h, four subsequent aircraft traverses were made along tracks C–D, E–F, G–H and I–J (at the times indicated in Fig. 3). These tracks were specified to approximate Lagrangian sampling of the air mass as it was transported to the East Coast.

Diagnostic air trajectories initialized at 1300 EST on

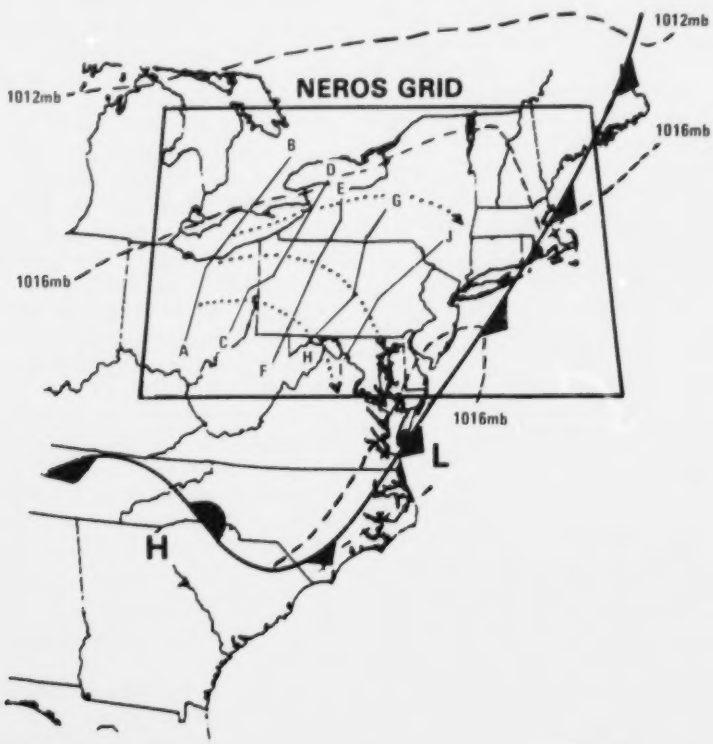
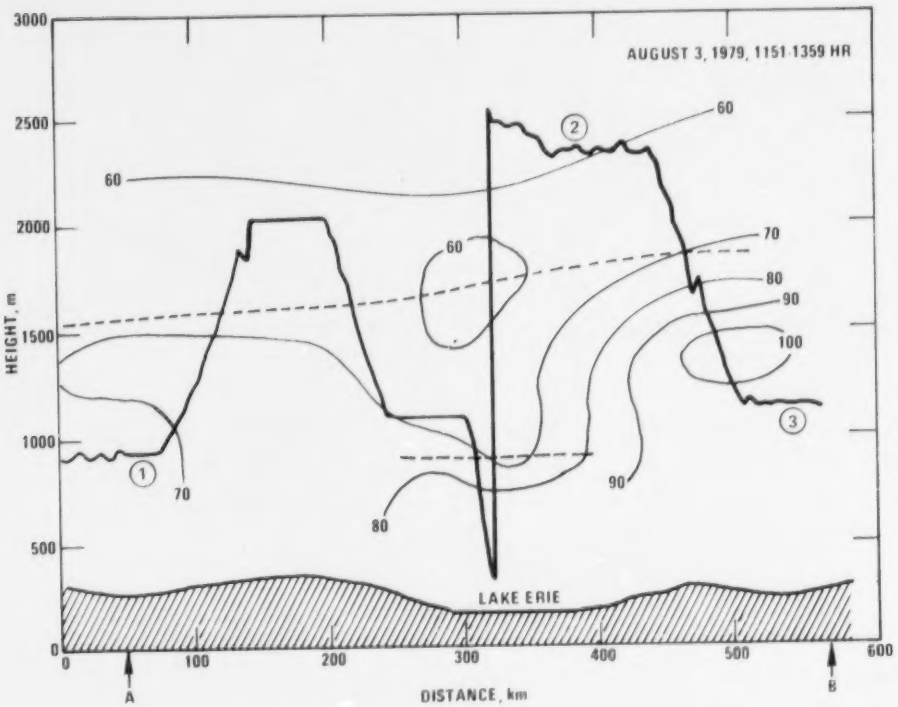
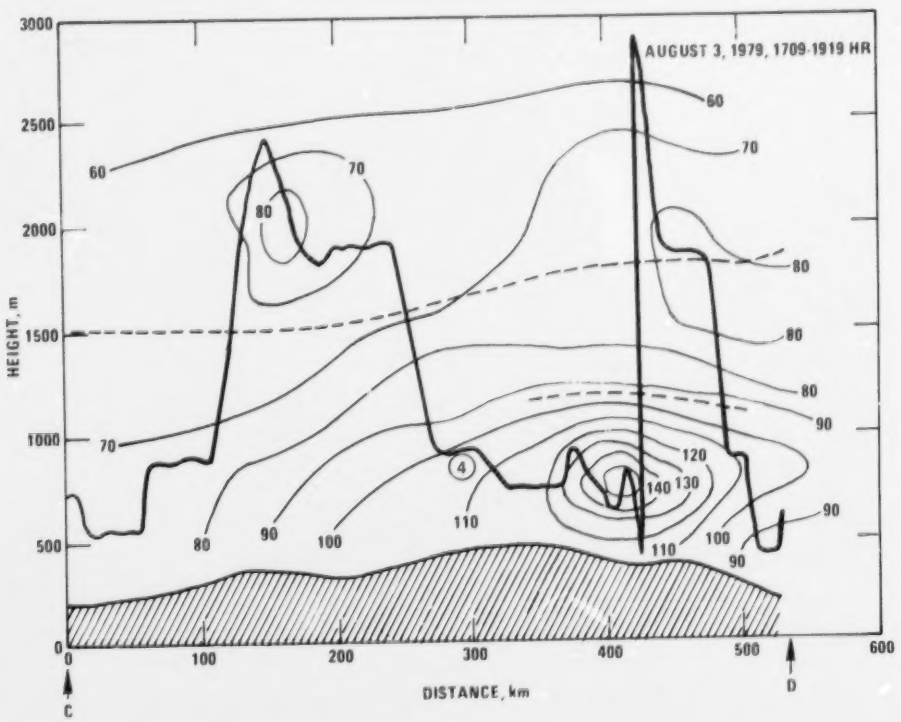



Fig. 1. Surface weather features and boundary-layer trajectories (dotted lines) for 3 August 1979, the NEROS modeling grid, and aircraft flight tracks for 3–4 August. Tracks are ~ 500 km except for G–H (400 km) and I–J, 450 km.

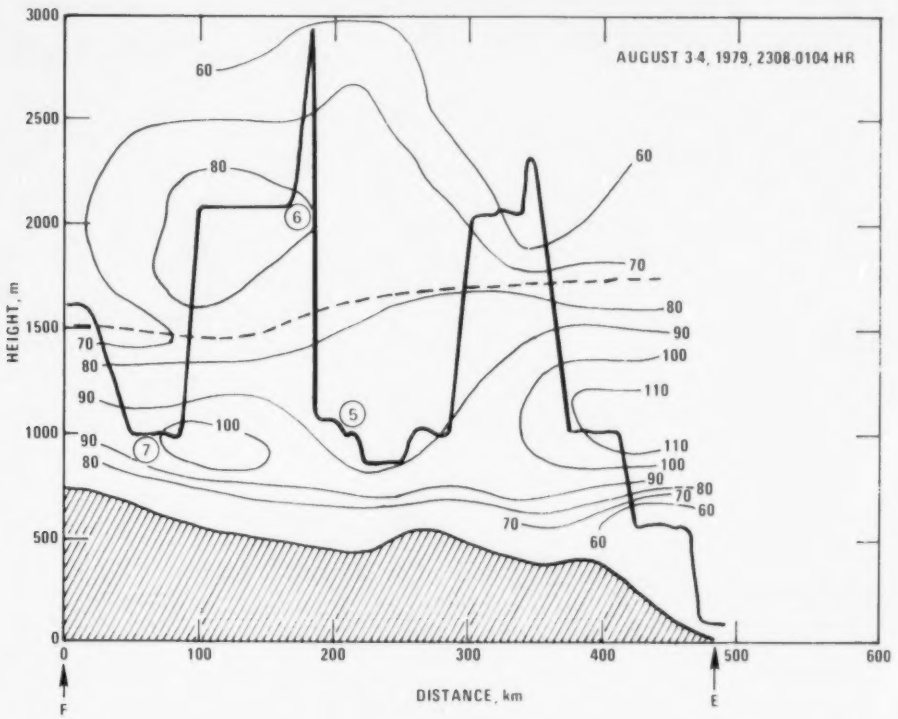

AUGUST 3, 1979, 1151-1359 HR
HEIGHT, m
DISTANCE, km
LAKE ERIE
A
B
1
2
3
60
70
80
90
100

(a)

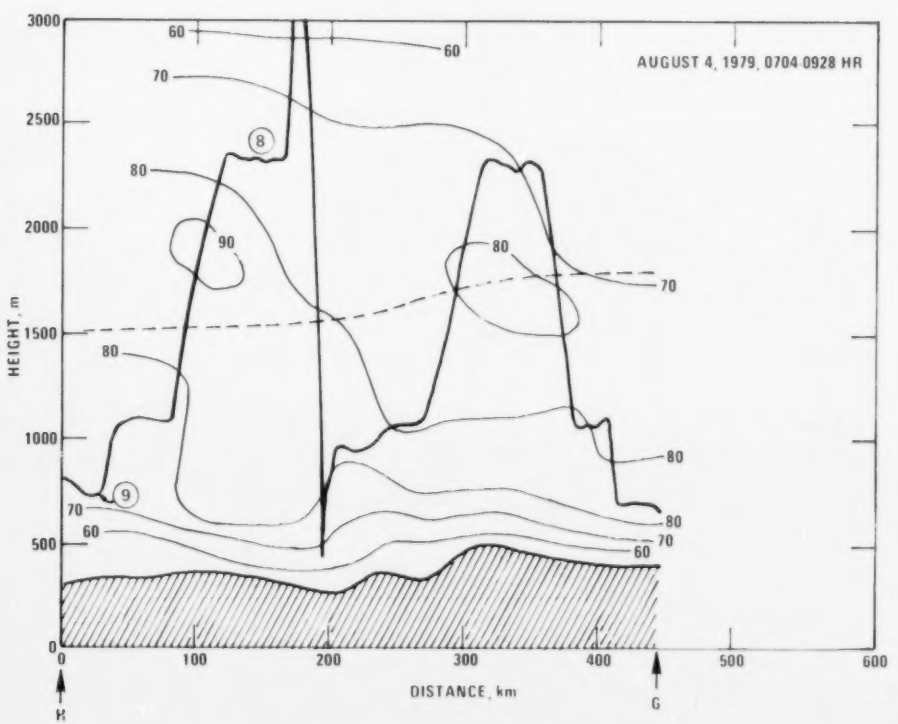

AUGUST 3, 1979, 1709-1919 HR
HEIGHT, m
DISTANCE, km
C
D
4
60
70
80
90
100
110
120
130
140

(b)

Fig. 2(a), (b)


AUGUST 3-4, 1979, 2308-0104 HR
HEIGHT, m
DISTANCE, km
F
E

(c)


AUGUST 4, 1979, 0704-0928 HR
HEIGHT, m
DISTANCE, km
H
G

(d)

Fig. 2(c), (d)

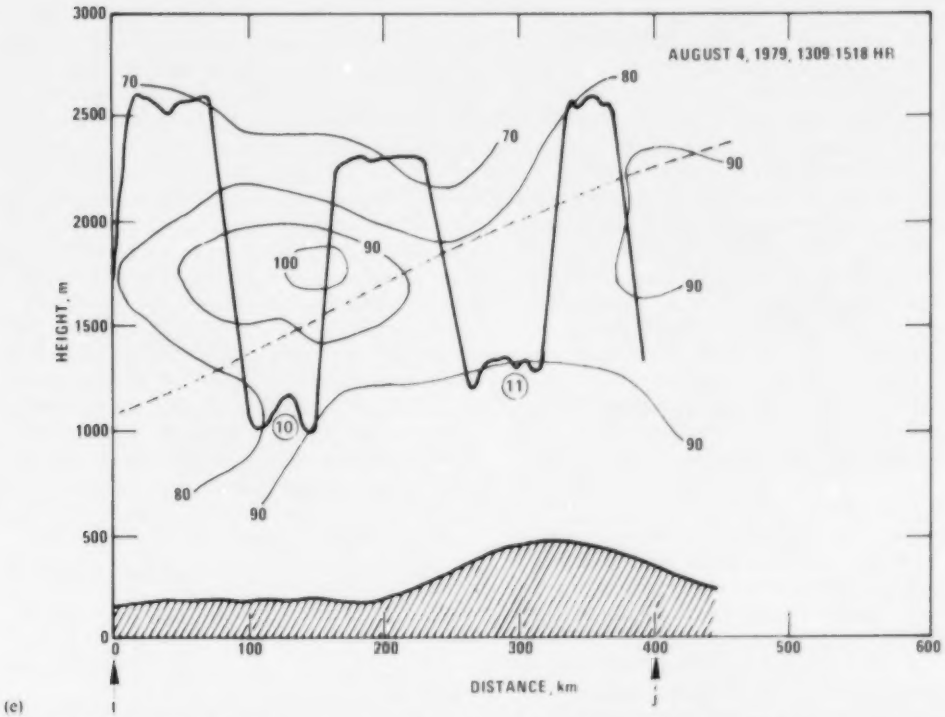



Fig. 2. Subjective analysis of ozone concentrations (ppb) measured along each flight traverse. The heavy solid lines indicate the aircraft path and thus the location of data points. The dashed lines indicate the top of the surface mixed layer (or the residual mixed layer). The circled numbers indicate the location of hydrocarbon samples. (a) Track A–B; (b) Track C–D; (c) Track E–F; (d) Track G–H and (e) Track I–J.

3 August at three points along track A–B are also shown in Fig. 1. The trajectories were computed using radiosonde data and the default option (winds averaged through the mixed layer) in the Heffter and Taylor (1975) trajectory model. The flights were generally east of the Lagrangian movement of the air mass as indicated by the diagnostic trajectories, especially in the southern part of the grid (see Fig. 3). The trajectories indicate divergence of the flow; air parcels originating in central Ohio moved to northern Virginia by 1900 EST the following day, while those over Lake Erie moved to southern New York. Divergence typically results in subsidence of the air mass and lowering of the boundary layer. In this case, however, the subsidence was probably compensated for by convective entrainment as there was no obvious lowering of the mixed layer height.

*Ozone concentration patterns*

The air-quality and meteorological data for each aircraft transect were subjectively analyzed to describe large-scale features along the transect. Analyzed $O_3$ cross sections for the five transects are shown in Fig. 2. The heavy solid lines represent the aircraft's location in the vertical plane of the transect and, thus, data points. The dashed lines represent the top of the mixed layer (or the residual mixed layer from the previous afternoon, for nocturnal transects). The height of the mixed layer was subjectively estimated from the aircraft profiles of $O_3$, $NO_x$, $\sigma_s$, temperature, humidity (diagrams similar to Fig. 2 were constructed for each of these parameters), and from National Weather Service radiosonde observations in the vicinity of the flight transects. The shaded area on the cross sections represents the average ground elevation along the ground track of the aircraft.

There were differences in the precision and accuracy of the measurement data from the three aircraft platforms. The differences, attributed to different instrumentation, sampling configurations, and procedures, were minimized in the analyses by adjusting the measured concentrations based on quality assurance procedures, audits, and intercomparison flights. The $O_3$ analyses in Figs 2(b) and (c) reflect a 15% downward adjustment of the Washington State University measurements and those in Figs 2(d) and (e) a 15% upward adjustment of the Brookhaven measurements.

$O_3$ measurements from surface monitoring sites near the flight tracks augmented the aircraft measurements in the analyses of Fig. 2. During daylight hours, $O_3$ concentrations at the surface monitoring sites

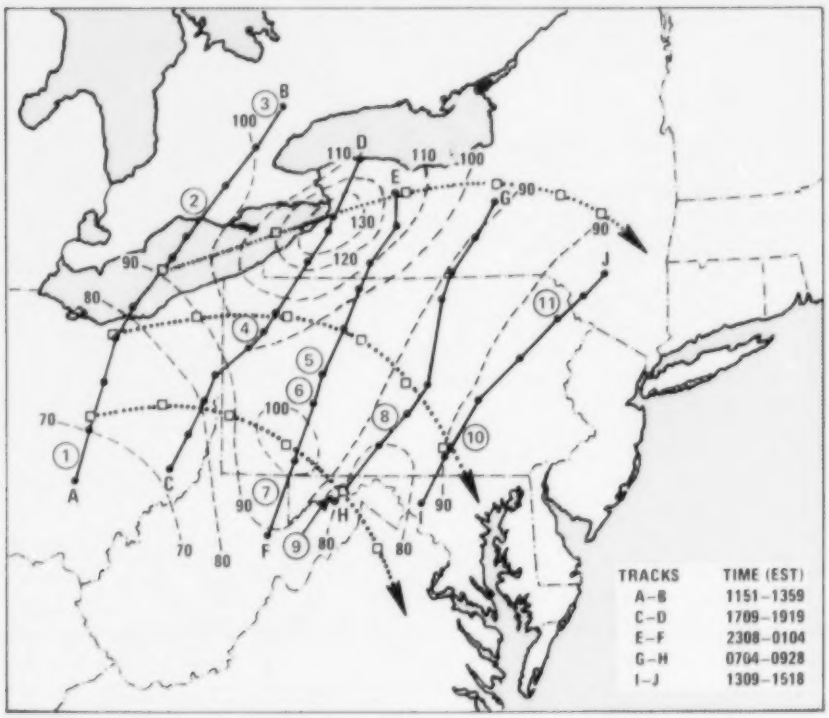



Fig. 3. Aircraft flight tracks, ozone concentrations (ppb) near the center of the mixed layer (dashed lines) and boundary-layer trajectories initialized along track A–B at 1300 EST on 3 August (dotted lines, the squares indicate 6 h positions). The circled numbers indicate the locations of hydrocarbon samples.

varied widely, particularly in the vicinity of urban areas, in response to varying local production and destruction processes. Therefore, the near-surface analyses in Figs 2(a), (b) and (e) should be regarded as highly idealized.

At night $O_3$ concentrations from surface monitoring sites were generally between 0 and 30 ppb, owing primarily to surface removal processes. The limited vertical resolution of the data in Figs 2(c) and (d) prevented an accurate analysis of the strong gradients that must exist due to the large differences in the $O_3$ observed at the surface and by the aircraft.

The analysis for the initial aircraft transect along track A–B, starting at 1151 EST on 3 August, is shown in Fig. 2(a). The mixing height was between 1500 m (in the south) and 1700 m MSL, except over Lake Erie, where it was less than 1000 m MSL. $O_3$ concentrations generally increased from south to north within the mixed layer, with high values observed over Lake Erie (>80 ppb) and in the northern part of the transect (>100 ppb). The horizontal gradient of $O_3$ above the mixed layer was weak; the analysis indicated a minimum of 55–60 ppb near the center of the track. Concentration patterns for $NO_x$ and $\sigma_s$ were similar, however, $\sigma_s$ values were highest in the southern part of the track (both within and above the mixed layer).

The $O_3$ analysis for the second transect (track C–D) is shown in Fig. 2(b). This track was the predicted 6-h displacement of the air mass sampled along track A–B. A plume with a maximum $O_3$ concentration near 160 ppb was observed on the northern part of the transect. A back trajectory analysis indicates the plume was over Lake Erie on the previous transect. Further, the analysis suggests the plume source to be from urban and industrial source complexes in the lower Great Lakes region. In-plume $O_3$ concentrations increased from 90 to 160 ppb while $NO_x$ concentrations decreased from 12 to 6 ppb and $\sigma_s$ increased from $3 \times 10^{-4}$ m$^{-1}$ to $6 \times 10^{-4}$ m$^{-1}$ from the first to the second transects. $O_3$ concentrations near 80 ppb were observed above the mixed layer in both northern and southern parts of this track.

Concentrations of $O_3$ associated with the plume on the third transect (Fig. 2c) were observed to be in excess of 110 ppb. However, this transect probably did not start far enough north to completely define the plume. The relatively low values of near-surface $O_3$ at the northern end of the transect may have been due to depletion of $O_3$ by fresh $NO_x$ emissions from the Buffalo area; $NO_x$ concentrations were observed by

aircraft to be in excess of 40 ppb near the surface shortly after take-off. Concentrations of $O_3$ in excess of 80 ppb were observed above the mixed layer in the southern part of the transects.

In contrast to the high $O_3$ concentrations persisting aloft in the residual mixed layer during this transect (and during transect G–H, Fig. 2d), the few $O_3$ measurements available from surface sites were between 0 and 30 ppb. Thus, the classical picture of $O_3$ being depleted in the surface layer at night while being transported aloft is supported by these data.

The fourth transect (Fig. 2d) was started at 0704 EST, 4 August out of Albany, NY. $O_3$ concentrations within the residual mixed layer were significantly smaller than for the previous transect. The observed differences may be due in part to sampling limitations; there are large areas on the cross section without observations (as on all the cross sections). High $O_3$ concentrations were measured above the mixed layer in both the northern and southern parts of the transect.

The final transect of the series (track I–J, Fig. 2e) was conducted in and above a well-mixed afternoon boundary layer upwind of the Northeast Corridor. Concentrations of $O_3$ were in excess of 80 ppb over most of the track, and high concentrations persisted above the mixed layer.

*Lagrangian ozone field*

Isopleths of $O_3$ concentration near the center of the mixed layer (or its nocturnal residual), extracted from the diagrams in Fig. 2, are shown in Fig. 3. The analysis represents a quasi-Lagrangian field in that changes in concentration along a trajectory (or an interpolated trajectory) roughly represents the Lagrangian derivative rather than a spatial change. $O_3$ concentrations were generally high over the grid, and the primary standard (0.12 ppm) was exceeded aloft over western New York. A back trajectory analysis suggests that the air mass over western New York at 04/1900 EST was over Chicago 36 h earlier and was transported over the conglomeration of urban source areas of northern Indiana and northern Ohio.

The latitudinal distribution of $O_3$ on the first two transects, see also Figs 2(a) and (b), generally coincided with the distribution of precursor sources (Clark, 1980), suggesting that the elevated concentrations were due to specific source areas rather than to a general air mass burden.

The temporal gradients of $O_3$ concentrations were consistent with general concepts of a Lagrangian budget. Between 1300 and 1800 EST on 3 August (tracks A–B and C–D, respectively), $O_3$ concentrations increased as expected from chemical production processes. The increase was largest in the northern portion of the grid (in the region of high precursor emissions). Concentrations generally decreased between 1800 and 0800 EST the following morning, and then increased on the afternoon 4 August (track I–J). Concentrations were generally lowest during the early-morning transect (track G–H).

The center trajectory in Fig. 3 is probably best suited for indicating the effect of regional $O_3$ transport, because it is free of the uncertainties associated with the ends of the flight tracks. $O_3$ concentrations along this trajectory, averaged from about 300 m above the surface to the top of the mixed layer (or the residual mixed layer), are plotted in Fig. 4 as a function of time. (The lower 300 m were excluded from the analysis because of the large uncertainty in the $O_3$ profile near the surface, especially at night.) The heavy lines link observations from the same aircraft; higher confidence can be placed in the relative change of these concentrations. $O_3$ concentrations along the trajectory increased between 1300 and 1800 EST and then remained constant or decreased over the next 12 h. Concentrations again increased between 0800 and 1400 EST on 4 August, being higher at 1400 over eastern Pennsylvania than at the same time the previous day over eastern Ohio.

$NO_x$ concentrations are also shown in Fig. 4. The uncertainty in the $NO_x$ measurements is greater than for $O_3$, due to the higher noise-to-signal ratio of the instrumentation. However, the temporal gradients of $NO_x$ and $O_3$ were generally consistent with those expected from photochemical processes (i.e. decreasing $NO_x$ with increasing $O_3$ production).

The temporal change of $O_3$ concentration (Fig. 4) along the center trajectory between 0800 and 1400 EST (4 August) was smaller than observed at surface sites over the same time period. The temporal change reflects mixing of the air aloft with relatively $O_3$-depleted surface air, and chemical production/removal processes. The net effect was an increase in $O_3$ concentration. The dashed line in Fig. 4 represents the temporal change in $O_3$ concentration average for two suburban surface sites located near the intersection of the trajectory with track I–J. (Note that the solid line represents Lagrangian measurements, and the dotted line Eulerian measurements.) The surface $O_3$ concentration increased from 10 to 90 ppb between 0800 and 1400 EST. Comparison of the two curves suggests that the increase may have been due mainly to fumigation of $O_3$ from the residual mixed layer aloft, rather than to local production.

*Ozone transport aloft*

Venting of pollutants above the boundary layer by cumulus clouds may be an important consideration in regional transport scenarios. In this process, clouds penetrate the top of the mixed layer, carrying boundary-layer air containing $O_3$ and other pollutants into a relatively stratified layer above. The cloud-vented pollutants are transported by a different wind environment than boundary-layer pollutants and eventually may reenter the boundary layer to impact the surface environment. The aircraft sampling plan included flights above the boundary layer to provide data for a qualitative assessment of vertical transport of $O_3$ and $O_3$ precursors due to cloud venting processes.

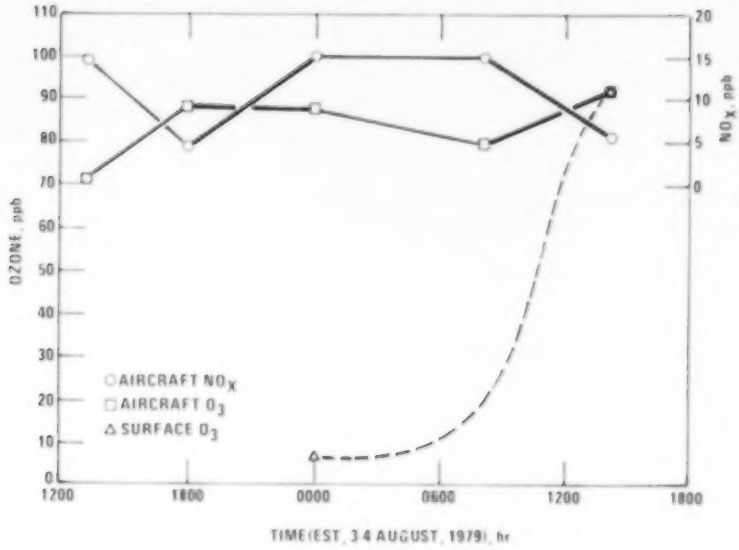



Fig. 4. Aircraft ozone (squares) and $NO_x$ (circles) concentrations (ppb) along the center trajectory of Fig. 3 and temporal change in surface concentration in eastern Pennsylvania (dashed line). The heavy lines link observations from the same aircraft.

Areas of relatively high concentrations of $O_3$ were observed above the mixed layer on most of the aircraft transects. As shown in Fig. 2(b), for example, $O_3$ concentrations near 80 ppb were observed above the mixed layer (over east central Ohio). The possible origin and fate of this high $O_3$ center were tracked through the use of an isentropic trajectory model (Haagenson and Shapiro, 1979). Figure 5 shows the 36-h backward and 24-h forward trajectories originating at 03/1900 EST at 800 mb (about 2000 m MSL) over east central Ohio. Cumulus clouds were observed along the back trajectory (areal coverage was reported to be almost 6/10 that afternoon), which supports cloud venting as the mechanism for transport of $O_3$ from within to above the mixed layer. High ozone concentrations were observed above the mixed layer on subsequent traverses, providing evidence of transport of the cloud vented material.

High $O_3$ concentrations above the mixed layer can result from intrusion of stratospheric air into the lower troposphere. However, the elevated $O_3$ values observed above the mixed layer in this study were

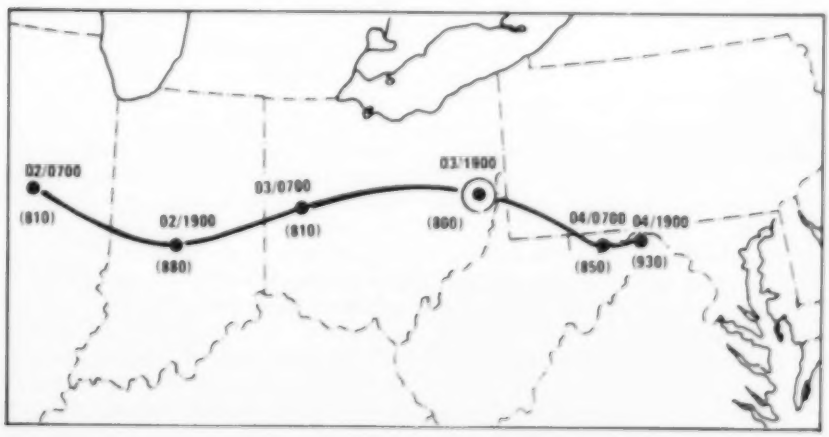



Fig. 5. Twelve-hour positions of forward and backward isentropic trajectories initialized at 03/1900 EST over east central Ohio. The numbers in parentheses are pressure (mb) for the indicated position.

accompanied by elevated $NO_x$ concentrations and $\sigma_\theta$ values suggesting an anthropogenic and thus, boundary layer origin.

The isentropic trajectory analysis (Fig. 5) suggests that the air did not remain above the mixed layer but apparently descended into the boundary layer on the afternoon of 4 August. The movement of air from above to within the boundary layer can occur through large-scale subsidence and subsequent entrainment into the boundary layer resulting from convective processes.

*Ozone production potential*

The chemical kinetics of $O_3$ production are very complex. However, the maximum potential for $O_3$ formation (under ideal solar radiation conditions) has been expressed as a function of nonmethane hydrocarbons (NMHC) and $NO_x$ through the Empirical Kinetic Modeling Approach (EKMA) (Dodge, 1977). EKMA results are specific to the modeled mixtures of both NMHC and $NO_x$, and are based on concentrations typical of urban areas. Concentrations of both NHMC and $NO_x$ measured in this study were generally low, often within the noise level of the instrumentation. Thus, the EKMA results reported here should be considered only qualitative at best.

Eleven grab (1–15 min.) samples were obtained by the aircraft during the 3–4 August experiment and later (within 48 h) analyzed for HC species. Total NMHC, $NO_x$, results of EKMA and other relevant information are given in Table 1. The location of the samples are indicated by the circled numbers in Figs 2 and 3.

Three HC samples were available for the first transect (track A–B); one at each end of the track within the mixed layer and one north of Lake Erie above the mixed layer. EKMA results indicate $O_3$ formation potential of about 10 ppb, which is not inconsistent with the increase in $O_3$ concentrations downwind of the second transect. Unfortunately a HC sample was not obtained in the mixed layer upwind of the high $O_3$ center observed over western New York on the second transect, i.e. over Lake Erie. The nocturnal hydrocarbon samples obtained in the previously mixed layer on tracks E–F and G–H indicate a residual potential for $O_3$ production of 15–55 ppb. Even in the residual cloud layer at night (locations 6 and 8, Table 1), there was potential for significant $O_3$ production. Finally, the air approaching the Northeast Corridor with $O_3$ concentrations of about 99 ppb (track I–J) appears to have an additional maximum potential of about 35 ppb. Thus, significant increases in ozone are possible even in the absence of new emissions.

## CONCLUSIONS

A high resolution data base was obtained in NEROS I and used for an evaluation of regional-scale transport of $O_3$. High concentrations of $O_3$ were observed on a regional scale. $O_3$ plumes within the air mass appeared to emanate from urban and industrial source area complexes. These plumes maintained some identity for travel times of the order of 12 h before blending with the air-mass burden.

A residual mixed layer aloft containing relatively high concentrations of $O_3$ was apparent during the nighttime traverses. $O_3$ concentrations in this layer did decrease during night-time transport but the decrease was small relative to the decrease in surface concentrations over the same period. Thus, relatively high concentrations of $O_3$ and $O_3$ precursors were transported aloft providing the potential for further $O_3$ production and impaction on surface concentrations after sunrise the following morning.

Lagrangian photochemical production of $O_3$ was observed during the day. However the production varied considerably over the air mass with the greatest production associated with precursor emissions from the urban and industrial source complexes of the lower Great Lakes region.

The study supports the idea that boundary layer $O_3$ and $O_3$ precursors may be vented above the mixed layer by cumulus activity, transported, and brought back into the mixed layer by subsidence and entrainment. Regional scale transport models should, therefore, include treatment of these processes.

Table 1. Total NMHC, $NO_x$ and ozone potential from EKMA based on hydrocarbon grab samples

| Track | Location | Altitude (m) | Time (EST) | NMHC (ppb) | $NO_x$ (ppb) | Ratio | $O_3$ Pot. (ppb) |
|---|---|---|---|---|---|---|---|
| A–B | 1 | 930 | 1258 | 13 | 16 | 0.8 | 8 |
| A–B | 2 | 2400 | 1428 | 7.4 | 9 | 0.8 | 8 |
| A–B | 3 | 1200 | 1451 | 22 | 16 | 1.4 | 10 |
| C–D | 4 | 895 | 1805 | 40 | 5 | 8 | 28 |
| E–F | 5 | 1049 | 0012 | 95 | 20 | 4.8 | 58 |
| E–F | 6 | 2072 | 0038 | 31 | 12 | 2.6 | 22 |
| E–F | 7 | 966 | 0053 | 28 | 18 | 1.6 | 16 |
| G–H | 8 | 2300 | 0852 | 57 | 20 | 2.8 | 36 |
| G–H | 9 | 760 | 0928 | 74 | 25 | 3.0 | 43 |
| I–J | 10 | 1100 | 1351 | 57 | 5 | 11.4 | 35 |
| I–J | 11 | 1300 | 1437 | 64 | 6 | 10.7 | 38 |

*Acknowledgements*—The authors greatly appreciate the outstanding performance and dedication of the personnel from Brookhaven National Laboratory, Washington State University and Research Triangle Institute who participated in the aerial sampling program. The authors also thank Phillip Haagenson for providing the isentropic trajectory analysis.

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